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Owner, by virtue of THE LAW הדין בעל ההמצאה מכח
an invention the title of which is ששמה הוא

(בעברית) (Hebrew)
חומרים משופרים מורכבים מפלסטיק ועץ

(באנגלית) (English)
IMPROVED WOOD-PLASTIC COMPOSITES

hereby apply for a patent to be granted to me in respect thereof. מבקש בזאת כי ינתן לי עליה פטנט

*בקשת חלוקה - Application of Division	*בקשת פטנט מוסף - Application for Patent Addition	*דרישה דין קדימה Priority Claim		
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חתימת המבקש Signature of Applicant *Luzzatto & Luzzatto* By: Attorneys for Applicant		היום 5 בחודש ינואר שנה 2003 This of the year לשימוש הלישכה		

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15167/02

חומרים משופרים מורכבים מפלסטיק ועץ

IMPROVED WOOD-PLASTIC COMPOSITES

IMPROVED WOOD-PLASTIC COMPOSITES

Field of the Invention

The present invention relates to the protection of wood-plastic composites from fungal attack. More particularly, the invention relates to the use of tetrabromobisphenol A and homologues and derivatives thereof in wood-plastic composites as a preservative against microorganism deterioration, particularly fungal attack.

Background of the Invention

In recent years, wood-plastic composites (WPCs) of many formulations have been developed to replace natural wood and their use is increasing at a significant rate annually. The most common types of wood-plastic composites are produced by mixing wood flour and plastics. (It should be noted that the terms "wood flour" and "wood particles" are used interchangeably throughout this specification to designate the wood component of the WPCs.) The resultant material can then be processed like plastics, for example extruded through a die to produce the final product. Many types of plastic are used including, for example, high and low density polyethylene, polypropylene, and PVC. The wood flour is typically made of recycled wood products, scrap wood, and sawdust. Several types of additives are commonly added to the mix, depending on

the type of material, production process, and end use of the final product. These additives include: coupling agents, to promote adhesion and dispersion of the particles of the mix; stabilizers, to prevent degradation during processing and service; UV stabilizers, to prevent degradation of the finished product; buffers; foaming agents, to reduce the density of the finished product; and lubricants, to improve flow and prevent edge damage in the extrusion process. Typical examples of formulations, production processes, and production equipment are given in the following U. S. Patents: US 6,344,504, US 6,180,257, US 6,117,924, US 5,981,631, and US 5,516,472.

Wood products in use and in storage are prone to deterioration by a variety of micro-organisms but especially fungi such as *poria placenta* and moulds. It is therefore common to use chemical preservative treatments to prevent such biological deterioration and there are many different wood preservatives known in the art. Typical of these are Creosote and Copper Chrome Arsenate. However, these preservative types display disadvantages such as high volatile organic compound (VOC) emissions (Creosote) and high heavy metal contents (CCA).

Articles produced from wood-plastic composites are less susceptible to biological attack than natural wood products because of the plastic which coats the wood particles. However at the edges of the article, wood

particles can be exposed to the surroundings and therefore the need for protection against attack does exist. It is not enough for the protection to be provided only on the surface of the article but it should also be provided for all of the wood particles in the wood-plastic composite material since any part of the item can become an outer surface during use when, for example a board is cut to its desired length with a saw. Since the composites are typically composed of approximately 50% each of wood particles and plastic, half of every exposed surface is susceptible to biological attack.

Tetrabromobisphenol A (hereinafter referred to as "TBBA") has been used, as described in JP 61-6769 (Publication No. 55-159915, dated December 12, 1980), to paint and coat a single plate of wood for the prevention of mould growth. Although the antifungal activity of TBBA has been known for at least 20 years it has not yet found practical application in industry.

It is therefore an object of this invention to provide wood-plastic composite materials that are resistant to biological wood deterioration.

It is yet another purpose of the invention to provide a method and compositions that do not require the use of harmful solvents.

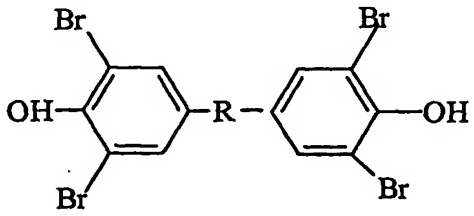
It is a further object of this invention to provide biocidal compositions based on TBBA, its homologues and derivatives that can be used to preserve wood-plastic composite materials in the absence of the disadvantages inherent in other preserving compounds.

It is a still further purpose of this invention to provide a method for the preservation of wood-plastic composite materials against fungal attack that employs the impregnation and/or mixing and/or coating and/or binding of the wood particles of the composite material with TBBA or its homologues and derivatives.

Further purposes and advantages of this invention will appear as the description proceeds.

Summary of the Invention

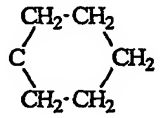
The present invention employs a biocidal composition comprising as its active ingredient Tetrabromobisphenol A (TBBA) [CAS RN = 79-94-7] or a homologue or a derivative thereof. TBBA is the tetrabrominated form of Bisphenol A of formula



Where, for TBBA, R is $C(CH_3)_2$.

By "homologues" of TBBA it is meant to indicate those compounds in which the Bisphenol A bridge is replaced by a different moiety. Illustrative and non-limitative examples of such homologues include:

- Tetrabromobisphenol F (TBBF), Bis(4-hydroxy-3,5-dibromophenyl)methane [CAS RN = 21825-03-6], R is CH_2;
- Tetrabromobisphenol Z (TBBZ), 4,4'-Cyclohexylidenebis(2,6-dibromophenol), [CAS RN = 53350-96-2], R is



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- Tetrabromobisphenol E (TBBE), 4,4'-Ethylidenebis(2,6-dibromophenol), [CAS RN = 126369-25-3], R is $CHCH_3$; and
- Tetrabromobisphenol S (TBBS), 4,4'-Sulfonyldi(2,6-dibromophenol), [CAS RN = 39635-79-5], R is SO_2.

By "derivatives" of TBBA it is meant to indicate those compounds that are further substituted by a substituent other than bromine, either on one or both phenyl rings, or at the bridge. Any such substitutions that do not

substantially alter the wood-preserving activity of the resulting compound with respect to TBBA are also encompassed by the present invention.

Preferably, the compound employed is TBBA in a solid state, such as powder or particles, or that has been solubilized in an organic or aqueous solvent. According to a preferred embodiment of the invention, the active compound is provided in aqueous solution. According to another preferred embodiment of the invention, the active compound is dissolved in an organic solvent such as alcohols, e.g. ethanol, hydrocarbons, toluene and ketones. According to still another preferred embodiment of the invention, the active compound is incorporated in an emulsion.

A biocidal wood preservative comprising TBBA as the active ingredient in aqueous solution can be solubilized, for instance, by the addition of TBBA to a solution comprising water, sodium hydroxide (NaOH), and sodium dithionite ($Na_2S_2O_4$). The concentration of TBBA in the solid state, aqueous solution, or organic solvent may be in the range of 0.01% (W/W) – 20% (W/W). More preferably, the concentration of TBBA may be in the range of 0.01% (W/W)-3% (W/W).

The long-term preservation, without mould growth or decay, of wood-plastic composite materials, is achieved by mixing and/or impregnating and/or coating the wood particles with an active ingredient, e.g., TBBA, a

derivative or a homologue of TBBA, or a mixture of two or more of the same, in solid form, an aqueous solution, an organic solvent, or in an emulsion before the addition of the other components during the process of the production of the WPC.

The method for applying the active ingredient to the wood particles comprises either impregnating the particles by pressure-treatment or, spraying the particles with, or soaking them in such a composition containing TBBA or its homologues and derivatives.

Detailed Description of Preferred Embodiments

The invention will now be described with reference to the following illustrative example, which is provided merely to illustrate the invention and is not intended to limit the scope of the invention in any manner.

The wood-plastic composite is comprised of a polypropylene (pp) homopolymer and wood flour in a weight ratio of about 50% pp: 50% wood. A lubricant, such as EBS wax, at a concentration of 1 to 2% of the total weight is added to these components.

In a preferred embodiment of the invention, before forming the composite material, a biocide comprising TBBA as the active ingredient in aqueous

solution is applied to the wood flour by, for example, mixing the dry TBBA with dry wood particles, vacuum impregnation, and/or coating.

It should be appreciated that in the case of large pieces of natural wood superficial coating of the wood with the active ingredient may not be sufficient to achieve long-term protection, and therefore in-depth impregnation of the wood with aqueous solution or organic solvent containing the active material, using various techniques, is preferred. The actual depth of penetration of the active material will depend on many factors, as will be apparent to the skilled person, such as the type of wood or wood product, the type of solution, the pressure employed, etc. However, at least some mixing of the wood with the active material, below the surface, must be achieved to insure improved protection. However, when particles of wood are employed surface coating, e.g., by spraying and uniting, with a TBBA composition is sufficient.

In the present invention it is also possible to employ vacuum impregnation. However long-term protection against fungal attack is provided, since the particles are first treated with the solution containing the active material and then dispersed essentially uniformly throughout the WPC material. For this reason, as well as the small size of the particles, it is sufficient to apply the active ingredient to the wood particles, by mixing with the solid active material, by soaking them in a

solution containing TBBA or its homologues and derivatives, or by spraying the particles with such a solution. After coating or impregnating the wood particles with the aqueous solution, they can be dried, either at ambient or at an elevated temperature, and stored for later use.

Preferably, the compound employed as the active ingredient is TBBA in solid form as a powder or particles or solubilized in an organic or aqueous solvent, or which is contained in an emulsion. According to a preferred embodiment of the invention, the active compound is provided in aqueous solution by the addition of TBBA to a solution comprising water, sodium hydroxide (NaOH), and sodium dithionite ($Na_2S_2O_4$). The concentration of TBBA (% by weight) may be in the range of 0.01% (W/W) – 20% (W/W). More preferably, the concentration of TBBA may be in the range of 0.01% (W/W)-3% (W/W).

According to another preferred embodiment of the invention, the active compound is dissolved in an organic solvent such as alcohols, e.g. ethanol, hydrocarbons, toluene and ketones.

The WPC is then manufactured according to procedures known in the art. For instance, if it is desired to employ an extruder for producing the wood-plastic composite material, the wood flour treated with TBBA is introduced into an extruder, e.g. a twin screw extruder, and dried at 200

to 240 degrees C. The polymer is metered into the twin screw extruder via a side single screw extruder at about 170 degrees C. At the same time, the lubricant is also metered into the twin screw. The wood, plastic, and lubricant are mixed and extruded through a die in the desired shape. The extruded product is cooled in a spray cooling tank.

Although embodiments of the invention have been described by way of illustration, it will be understood that the invention may be carried out with many variations, modifications, and adaptations, without departing from its spirit or exceeding the scope of the claims.

Claims

1. An improved wood-plastic composite (WPC) produced from a mixture comprising, wood particles, plastic, and optionally additives, and further comprising an active ingredient that acts as a biocide, and which comprises TBBA or a homologue or derivative thereof, wherein said active ingredient is added to said mixture together with said wood particles during and/or before production of the WPC.

2. An improved WPC according to claim 1, wherein the active ingredient is added to the mixture together with the wood particles according to one or more methods chosen from the following group:

- impregnation of said wood particles with said active ingredient;
- coating of said wood particles with a solution or emulsion containing said active ingredient;
- soaking said wood particles in a solution or emulsion containing said active ingredient; and
- mixing said wood particles together with said active ingredient in particulate and/or powder form.

3. An improved WPC according to claim 1, wherein the coating of the wood particles with a solution or emulsion containing the active ingredient is accomplished by spraying said wood particles with said solution or emulsion.

4. An improved WPC according to claim 1, wherein the active ingredient is Tetrabromobisphenol A (TBBA) or a homologue or a derivative thereof.

5. An improved WPC according to claim 1, wherein the active ingredient is a solid.

6. An improved WPC according to claim 1, wherein the active ingredient is dissolved in a solvent.

7. An improved WPC according to claim 6, wherein the solvent is an aqueous solvent.

8. An improved WPC according to claim 7, wherein the aqueous solvent comprises water, sodium hydroxide (NaOH), and sodium dithionite ($Na_2S_2O_4$).

9. An improved WPC according to claim 6, wherein the solvent is an organic solvent.

10. An improved WPC according to claim 9, wherein the organic solvent is selected from alcohols, e.g. ethanol, hydrocarbons, toluene and ketones.

11. An improved WPC according to claim 6, wherein the active ingredient is TBBA at a concentration in solution in the range of 0.01% (W/W) to 20%(W/W).

12. A method of producing an improved wood-plastic composite (WPC) containing an active ingredient that acts as a biocide comprising mixing particles or a powder of said active ingredient with the wood particles or impregnating and/or coating said wood particles with said active ingredient by means of vacuum impregnation, soaking them in a solution or emulsion containing said active ingredient, or by spraying the particles with such a solution or emulsion before combining them with the plastic component and optional additives from which the WPC is produced.

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